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THITHER: PHOTOCURING PRESSURE-SENSITIVE ADHESIVE
COMPOSITION

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Amendments: There are no amendments to this patent.

[note: All names, addresses, company names, and brand names are translated in the most common manner. Japanese language does not have singular or plural words unless otherwise specified with numeral prefix or general form of plurality suffix. The title of this patent is translated as photocuring; however, it may be also translated as light curing as well. translator's note]

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TITLE OF THE INVENTION: Photocuring pressure-sensitive adhesive composition

CLAIMS

1. A photocuring pressure-sensitive adhesive composition has characteristics as such that contains following components:

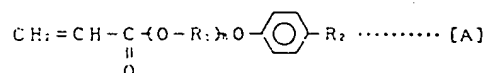
I. 30 to 50 parts by weight acryl group copolymer showing 50,000 to 150,000 weight average molecular weight prepared through copolymerization of monomer mixture containing 0.5 to 10 parts by weight (c) shown below based on total 100 parts by weight (a) and (b) components shown below.

(a) component: alkyl ester acrylate or alkyl ester methacrylate,

(b) component: polymerizable unsaturated monomer having polar groups

(c) component: polymerizable unsaturated monomer having functional groups with photosensitizing action

II. 40 to 60 parts by weight monofunctional acryl group monomer shown with following general formula [A]:



(In above formula, R₁ shows alkylene group with 1 to 3 carbon atoms; R₂ shows hydrogen or alkyl group with 1 to 15 carbon atoms; and n shows 1 to 10 integers.)

III. 5 to 15 parts by weight tackifier resin

IV. photoinitiator

DETAILED EXPLANATION OF THE INVENTION

[Field of industrial application]

The present invention relates to a photocuring pressure-sensitive adhesive composition with excellent screen printing aptitude.

[Prior art and problem points]

According to the cases of flexible printed circuit assembly, or combining said printed circuit boards with various electrical and electronic machinery and tools, practice to paste said printed circuit boards mutually or to paste printed circuit board with other substrate is implemented. In addition, such pasting practice is generally conducted through inclusion of a double coated pressure-sensitive adhesive sheet, screen printing thermosetting pressure-sensitive adhesive agent where pasting of printed circuit board is required and forming a tacky adhesive layer directly on said printed circuit board.

However, according to the method that uses double coated pressure-sensitive adhesive sheet, it requires many processes such as a process to stamp out according to the shape of portion requiring pasting, a process to remove a liner from one plane of pressure-sensitive adhesive sheet, pasting after position is matched, and a process to remove remaining liner of pressure-sensitive adhesive sheet to present problem points of poor productivity. In addition, this method requires preparation of double coated pressure-sensitive adhesive sheet prior to pasting to require production equipment and material for such purpose to result in problem point of high cost.

In addition, according to the method that uses thermosetting pressure-sensitive adhesive agent, because pasting is conducted through forming a tacky adhesive layer directly on a printed circuit board, it displays excellent productivity as well as economy; however, because film substrate of printed circuit board is generally constructed of polyester that shows poor heat resistance, it is difficult to apply sufficient curing conditions (sufficient baking temperature and sufficient baking time) to said pressure-sensitive adhesive agent to tend to lack curing of tacky adhesive layer to consequently present problem points of lack of pressure-sensitive adhesive strength or oozing of pressure-sensitive adhesive agent from surroundings of pasted part.

According to the photocuring pressure-sensitive adhesive agent that was developed recently and is commercialized in some areas, it does not require processes such as heating at all, and it sufficiently cures through only light irradiation of as short time as several seconds to several tens of seconds; and therefore, it can prevent from occurrence of problem points such as lack of adhesive strength or oozing of adhesive agent which occur in the case of above-explained thermosetting pressure-sensitive adhesive agent, and is consequently suited for pasting and the like of flexible printed circuit boards.

However, according to above-explained conventional photocuring pressure-sensitive adhesive agent, for instance, as disclosed in the Japanese Patent Application Kokai Sho 60[1985]-99183 specification, pressure-sensitive adhesive agent comprising acryl copolymer to which functional groups with photosensitization action are induced, and many of other types of pressure-sensitive adhesive agents are all known to show significantly high viscosity to present a large problem point that screen printing method cannot be used for forming of tacky adhesive layer. In particular, as in the case of assembly of printed circuit board, not being able to use screen printing to form a tacky adhesive layer to only the portions requiring adhesion displays not only very critical defects, but also significant hindrance on the productivity. And therefore, in the case of screen printing of conventional photocuring pressure-sensitive adhesive agent, it is generally practiced to add dilutant that is either reactive or nonreactive with said adhesive agent to dilute to viscosity that is suited for screen printing; however, when reactive dilutant is used, it causes collapse of balance of pressure-sensitive adhesive characteristics including cohesive force of tacky adhesive layer or peel strength; and in addition, when nonreactive dilutant is used, it not only results in difficult film thickness control of adhesive layer due to decline in concentration of adhesive agent, but also heating process may be required in order to remove nonreactive dilutant in some cases to present many problem points which need modifications.

That is to say, the purpose of the present invention is to solve above-explained problem points of conventional photocuring adhesive agent by offering a photocuring pressure-sensitive adhesive composition (this will be hereafter simply referred to as pressure-sensitive adhesive agent) with characteristics of containing following components:

(a) component: alkyl ester acrylate or alkyl ester methacrylate

(c) component: polymerizable unsaturated monomer having functional groups with photosensitization action.

$$\text{CH}_2=\underset{\text{H}}{\underset{\text{C}}{\text{C}}}-\text{O}-\text{O}-\text{R}+\text{O}-\text{C}_6\text{H}_4-\text{R} \cdots \cdots (\text{A})$$

(In above formula, R₁ shows alkylene group with 1 to 3 carbon atoms; and R₂ shows hydrogen or alkyl group with 1 to 15 carbon atoms; and n shows 1 to 10 integers.)

III. 5 to 15 parts by weight tackifier

IV. photoinitiator

As for above-explained acryl copolymer [I] used in this invention, the (a) component that constructs said copolymer includes alkyl ester acrylate or alkyl ester methacrylate, and concrete examples include alkyl ester acrylates such as ethyl acrylate, butyl acrylate, 2-ethyl hexyl acrylate, octyl acrylate; and alkyl ester methacrylates such as methyl methacrylate, ethyl methacrylate, and butyl methacrylate, and these esters may be used either alone or as mixture of more than two types. In addition, it is all right to jointly use part of other unsaturated monomers, for instance, vinyl group monomers such as vinyl acetate or styrene which are copolymerizable with these esters.

As for the polymerizable unsaturated monomer having polar group that is the (b) component shown in above-explained acryl copolymer [I], unsaturated monomers having polar groups such as carboxyl group, hydroxyl group, or amino group may be mentioned; and in particular, acryl group unsaturated monomers having these polar groups are recommended; and their concrete examples include acrylic acid, methacrylic acid, 2-hydroxyethyl ester methacrylate, and diethyl amino ethyl ester acrylate.

As for the polymerizable unsaturated monomer having functional groups with photosensitization action of the (c) component in above-explained acryl group copolymer [I], for instance, following may be mentioned:

(1) Monomers disclosed in the USA Patents 3,004,073 and 3,575,925 may be used; and one examples includes the one that is synthesized by inducing carboxyl group or hydroxyl group to benzophenone, benzoin, or anthraquinone, and by reacting this with glycidyl (meth)acrylate, in other words, it includes 2-acryloyloxy benzophenone, 1-acryloyloxy-2-[4-(p-chlorobenzoyl)benzoyloxy]ethane.

(2) Reaction products of polychloro phenyl glycidyl ether and (meth)acrylic acid.

(3) 3-(pentachloro phenoxy)-2-hydroxy propyl methacrylate

According to this invention, because acryl copolymer [I] comprising above-explained (a) through (c) components is used as a constitutional component of pressure-sensitive adhesive agent, when said pressure-sensitive adhesive agent is coated and is irradiated with light, it is estimated that the functional groups with photosensitization action based on the (c) component of the copolymer [I] reacts with monofunctional acryl monomer [II] that is a separate constitutional component to form macromolecules to form homogeneous crosslinking bonding that proceeds at the same time; and as a result, tacky adhesive layer showing excellent pressure-sensitive adhesive characteristics is formed.

And therefore, according to the acryl copolymer [I] used in this invention, because copolymerization ratio of above-explained (a) through (c) components, or in particular, copolymerization ratio of the component (c), is the important factor that affects pressure-sensitive adhesive characteristics, said copolymerization ratio must be as such that the (c) component is within a range of 0.5 to 10 parts by weight based on total 100 parts by weight (a) component and (b) component; and it is more preferable when it is within a range of 1 to 6 parts by weight. When copolymerization ratio of (c) component happens to be less than 0.5 parts, pressure-sensitive adhesive characteristics of given pressure-sensitive adhesive composition become poor; and on the other hand, when it happens to be greater than 10 parts by weight, crosslinking bonding is formed at excess level in the tacky adhesive layer to cause too strong cohesive force to result in poor adhesive force.

Furthermore, according to this invention, although there is no special restriction placed on the copolymerization ratio of (a) component and (b) component, and may be set at optional rate, it is recommended to set within such range of (a) component : (b) component as 90 to 99.5 : 0.5 to 10 weight % from the standpoint of improvement on adhesive force to either substrate or adherend.

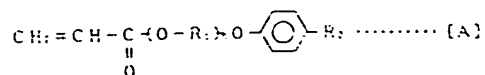
It is necessary that the weight average molecular weight (this term will be hereafter abbreviated as Mw) of the acryl copolymer [I] comprising above-explained components (a) through (c) is within a range of 50,000 to 150,000; and when copolymer [I] of which Mw escapes above-explained range is used, that is to say, when Mw happens to be smaller than 50,000, cohesive force after light irradiation remains small to provide only poor pressure-sensitive adhesive characteristics; and in addition, when Mw happens to be beyond 150,000, it significantly raises viscosity of pressure-sensitive adhesive agent to cause plate peeling during screen printing, foaming, and poor leveling to show poor printing aptitude.

No special methods are required in producing acryl copolymer [I] comprising above-explained (a) through (c) components; and various already known polymerization methods may be used. One preferred example method includes solution polymerization method that carries out polymerization reaction in organic solvents such as toluene, ethyl acetate, or methyl ethyl ketone; and during this solution polymerization, peroxides such as benzoyl peroxide or lauroyl peroxide, or azo compound such as azobisisobutyronitrile may be used as polymerization initiators; and in addition, molecular weight regulators such as alpha-methyl styrene and the like may be used as needed; and copolymer with subjected Mw may be prepared through adjustment of application rate, polymerization reaction temperature, and polymerization time of these polymerization initiators in appropriate manner.

As for above-explained acryl copolymer [I], it may be used within a range of 30 to 50 parts by weight in this invention's pressure-sensitive adhesive composition. When constitutional ratio of said copolymer [I] happens to be less than 30 parts by weight, surface pressure-sensitive adhesive force of given pressure-sensitive adhesive agent remains poor; and when it happens to exceed 50 parts by weight, viscosity of given adhesive composition becomes high to show poor screen printing aptitude.

Furthermore, when acryl copolymer [I] is prepared through said solution polymerization method, because said copolymer [I] is in a form of polymer solution containing organic solvents, it is recommended to remove the solvents from this polymer solution. Removal of solvents may be conducted any optional timings such as after completion of solution polymerization, during compounding of pressure-sensitive adhesive agent, after compounding of said agent, by heating or reducing the pressure of polymer solution, or combination of heating and pressure reduction.

According to the monofunctional acryl monomer [II] used in this invention, it refers to the compound shown with general formula [A] below; and said compound is set with viscosity suited for screen printing without inhibiting pressure-sensitive adhesive characteristics of given pressure-sensitive adhesive agent at all; and this is an important component as reactive dilutant in this invention.



(In above formula, R₁ shows alkylene group with 1 to 3 carbon atoms; and R₂ shows hydrogen or alkyl group with 1 to 15 carbon atoms; and n shows 1 to 10 integers.)

Concrete examples of monofunctional acryl monomer [II] used preferably in this invention include phenoxy ethyl acrylate, phenoxy propyl acrylate, nonyl phenoxy ethyl acrylate, nonyl phenoxy propyl acrylate and the like.

Above-explained monofunctional acryl monomer [II] may be used at such constitutional ratio of within a range of 40 to 50 parts by weight in this invention's pressure-sensitive adhesive composition. According to said constitutional ratio, when it happens to be less than 40 parts by weight, viscosity of adhesive agent becomes too high to result in poor screen printing aptitude; and on the other hand, when it happens to exceed 60 parts by weight, cohesive force and peel force of given tacky adhesive layer become poor, and is not desirable.

Furthermore, according to this invention, it is also useful to use other reactive dilutants jointly with above-explained acryl monomer [II] with purpose of further improving pressure-sensitive adhesive characteristics such as heat resistance, cohesive force, and adhesive force of given adhesive composition; and in particular, compounds having more than two acryloyl groups in one molecule, for instance, reactive dilutants comprising polyfunctional (meth)acrylate compounds such as 1,4-butane diol diacrylate, diethylene glycol diacrylate, or trimethylol propane triacrylate are recommended for use. However, compounding of large amount of said reactive dilutants may cause collapse of balance in pressure-sensitive adhesive characteristics; and therefore, it is recommended to use within a range of 0.5 to 2 weight % in the pressure-sensitive adhesive composition.

The tackifier resin used in this invention, through combination with above-explained acryl group copolymer [I] and monofunctional acryl group monomer [II], contributes toward significant improvement on peel strength of final pressure-sensitive adhesive agent product; and this tackifier resin may be appropriately selected and used so it would provide good miscibility and solubility mutually with acryl group copolymer [I] and monofunctional acryl group monomer [II]. As for the preferred tackifier resins, rosin modified resin, terpene modified resin, and petroleum oil resin may be mentioned.

Above-explained tackifier should be within a range of 5 to 15 parts by weight constitutional ratio of this invention's pressure-sensitive adhesive composition; and when this constitutional ratio happens to be less than 5 parts by weight, it shows poor improved effect of peel strength; and when it happens to exceed 15 parts by weight, despite of improved peel strength, tacky adhesive layer remains hard to show poor surface pressure-sensitive adhesive force and heat resistance adhesive properties.

According to this invention's photocuring pressure-sensitive adhesive composition, because its constitutional component is of acryl copolymer [I] with induced functional groups showing photosensitization action, a photocuring pressure-sensitive adhesive agent showing excellent pressure-sensitive adhesive characteristics may be given even when it is constituted of said copolymer [I], monofunctional acryl monomer [II], and tackifier only; however, curing speed by light irradiation remains fairly slow; and therefore, it is not suited for improvement of productivity that is the purpose of this invention. And therefore, photo initiator is used further as another separate component in this invention. The photoinitiator that is used is not particularly restricted; and for instance, photoinitiators of cleavage type such as benzoin ethyl ether, benzyl dimethyl ketal, or diethoxy acetophenone; or photoinitiators of hydrogen extraction type such as benzyl, benzophenone, or 2,4-diethyl thio xanthone may be used favorably; and it is recommended to set the application rate of these photoinitiators to within a range of 0.1 to 4 weight % based on the adhesive composition.

This invention's adhesive composition having a constitution explained above may be formed as an adhesive layer showing excellent pressure-sensitive adhesive characteristics through screen printing by using appropriate screen printing plate on optional substrate and irradiation of light. It is preferable when light source used for said light irradiation is generally of UV ray with 180 to 400 nm range of wavelength; and use of high pressure mercury lamp or metal halide lamp is recommended; and irradiation time of light is appropriate in the case of high pressure mercury lamp with 80 W/cm², 6 to 15 seconds with energy dosage of 300 to 800 mJ/cm² is generally appropriate.

A case of using this invention's photocuring pressure-sensitive adhesive composition for flexible printed circuit board through screen printing method is explained above; however, this invention should not be limited to this; and for instance, said pressure-sensitive adhesive agent may be coated on various substrate through already known coating methods such as roller coating, curtain flow coating, spray coating, or gravure coating; and it goes without saying that through such process, it may be used for various applications including double coated pressure-sensitive adhesive tapes, pressure-sensitive adhesive labels, and pressure-sensitive adhesive sheets.

[Examples]

Examples of this invention are explained below. In the examples, the term "parts" used in the compounding composition refers to parts by weight.

EXAMPLE-1

65 parts of 2-ethyl hexyl acrylate, 20 parts of butyl acrylate, 5 parts of vinyl acetate, 5 parts of acrylic acid, 5 parts of 1-acryloyloxy-2-[4-(pchlorobenzoyl) benzoyloxy] ethane, and 233 parts of ethyl acetate were placed in a reactor substituted with inert gas; and then, 0.5 parts of azobis isobutyronitrile was used as a polymerization initiator to subject this to a solution polymerization to give ethyl acetate solution of acryl group copolymer [I] with 100,000 weight average molecular weight.

To 133 parts of this copolymer solution, 48 parts of nonyl phenoxy ethyl acrylate, 12 parts of terpene phenol resin, 1 part of 1,4-butane diol diacrylate, and 2 parts of benzyl dimethyl ketal were mixed; and in addition, hydroquinone monomethyl ether with 200 ppm was added as a heat polymerization inhibitor; and this was raised to about 70°C temperature, and ethyl acetate in the solution was removed under reduced pressure to give a photocuring pressure-sensitive adhesive composition with 100 poise viscosity at 25°C.

The photocuring pressure-sensitive adhesive composition prepared in above-explained manner was used to screen print on a polyester film to give 30 μ m film thickness by using a screen printing late with 150 mesh and bias weave; and this was irradiated with light from two high pressure mercury lamps (80W/cm) installed at 15 cm above this printing plane for 7 seconds to cure adhesive layer to give a pressure-sensitive adhesive sheet. The adhesive sheet given in this manner was evaluated for its pressure-sensitive adhesive characteristics based on the test methods explained below; and in addition, during above-explained screen printing operation, screen printing aptitude including plate separation state (plate separation) of adhesive agent from screen printing plate, state of foam involvement (foaming feature) on adhesive layer, and flatness (leveling feature) of adhesive layer surface were evaluated. Results of each evaluation are shown in Table-1.

TESTING METHODS:

(1) 180° peel force: A pressure-sensitive adhesive sheet is cut in a tape form 25 mm width; and this tape was press adhered to a stainless sheet by moving 2 kg roller in one complete back and forth motion; and this was measured for 180° peel force by pulling with tensilometer after 30 minutes of said press adhesion at 23°C at 300 mm/minute tensile speed.

(2) 40° retention force: A pressure-sensitive adhesive sheet is cut in a tape form with 25 mm width; and one end of this tape (25 mm) is pasted to a stainless sheet; and 1 kg load is applied to the other end of the tape to hold this tape in this state at 40°C x 20 hours to measure sliding distance of this tape.

(3) Ball tack : A pressure-sensitive adhesive sheet is pasted on a flat plate with 30° incline with its adhesive layer facing upward over 10 cm; and furthermore, stainless ball is rolled from 10 cm runway distance; and surface pressure-sensitive adhesive force is shown with the size (ball No.) of ball where it stopped on the tacky adhesive layer. Furthermore, atmospheric temperature during the test is set at 23°C.

EXAMPLES - 2 THROUGH 5

Photocuring pressure-sensitive adhesive compositions were adjusted in the same manner as explained in the example-1 by using each component explained in said example-1 according to the prescriptions shown in the Table-1; and they were evaluated in the same manner. Evaluation results are jointly shown in the Table-1.

COMPARATIVE EXAMPLES - 1 THROUGH 9

Photocuring pressure-sensitive adhesive compositions were adjusted in the same manner as explained in the example-1 by using each component explained in said example 1 according to the prescriptions shown in the Table-1; and they were evaluated in the same manner. Evaluation results are shown in the Table-1.

Table-1

A: examples, comparative examples,
B: adhesive compositions, evaluation results,
C: examples, D: comparative examples

I: acryl copolymer [I], Ia: ACBE compounding rate (*1), Ib: Mw ($\times 10^2$) of said copolymer,
II: compounding composition of pressure-sensitive adhesive agent (parts), IIa: acryl copolymer [I], IIB: monofunctional acryl monomer [II], IIC: tackifier resin
III: viscosity of adhesive agent (poise / 25°C),
IV: evaluation results, IVa: screen printing aptitude (*2),
a: plate separation, 2: foaming, 3: leveling feature, IVb: pressure-sensitive adhesive characteristics, a: 180° peel force (kg/25 mm), 2: 40°C holding force (*3), 3: ball tack 4: minutes

E: Explanation of Tables

F: (*a) ACBE: 1. acryloyloxy-2-[4-(p-chlorbenzoyl) benzoyloxy] ethane,

G: (*2) explanation of codes

plate separation o; good plate separation, x; not possible screen printing,

foaming feature o; no foaming on printed plane, good result, x; not possible screen printing

leveling feature o; flat and smooth printed plane with good leveling feature, x; not possible screen printing

H: (83) explanation of codes

NC: absolutely no sliding, very good result, 3 mm; sliding distance is 3 mm and lacks holding force, 20 minutes; pressure-sensitive adhesive sheet fell off from stainless after 20 minutes to indicate very poor holding force,

比較例・実例	C 実 例					D 比 較 例								
	1	2	3	4	5	1	2	3	4	5	6	7	8	9
アクリル共重合体 (I) Ia	5	5	5	5	5	12	5	5	5	5	5	5	5	5
ACBE配合率(%) Ib	100	100	70	100	100	100	100	100	100	40	200	100	100	50
圧着剤の配合組成 (II) IIa	45	35	45	40	45	45	45	20	60	45	45	40	40	100
アクリル共重合体 (I) Ib	43	53	43	47	43	43	43	68	38	43	43	50	40	-
圧着剤の配合組成 (II) Ic	12	12	12	9	12	12	12	12	12	12	12	-	20	-
粘度 (mPa・s / 25°C) III	100	75	20	95	100	100	110	20	200	60	100	105	95	200
スクリーン印刷適性 (IV) IVa	o	o	o	o	o	o	o	o	x	o	x	o	o	x
板剥離性 (IVb) a	o	o	o	o	o	o	o	o	x	o	x	o	o	x
泡盛性 (IVb) 2	o	o	o	o	o	o	o	o	x	o	x	o	o	x
レベリング性 (IVb) 3	o	o	o	o	o	o	o	o	x	o	x	o	o	x
圧着剤の配合組成 (II) IVb	1200	1500	1500	1500	2000	1800	1200	1200	-	1200	-	2000	2000	-
40°C保持力 (kg/25mm) 2	NC	NC	NC	NC	NC	NC	NC	NC	-	3	-	NC	200	-
ボールタック (3) 3	6	5	5	7	4	3	1	1	-	2	-	4	1	-

E: 説明

F: (*a) ACBE: 1. アクリロイルオキシ-2-[4-(p-クロルベンゾイル) ベンゾイルオキシ] エタン,

G: (*2) 説明: o: 剥離可能, x: スクリーン印刷不能,

泡盛性: o: 印刷面に泡盛はみられない, x: スクリーン印刷不能,

レベリング性: o: 印刷面が平滑でレベリング性良好, x: スクリーン印刷不能,

H: (83) 説明: NC: 全く滑らず, 3mm: 滑り距離3mmで保持力に落ちる, 20分: 貼付後20分経過後に剥離したとき, 剥離力弱くて悪い,

[Effects of this invention]

The photocuring pressure-sensitive adhesive composition of this invention uses a base that is composed of acryl copolymer with specific composition to which functional groups with photosensitization action are induced; and acryl monomer with specified structural formula is added to this as a reaction dilutant; and in addition, specified rates of tackifier and photoinitiator are compounded; and therefore, it shows excellent screen printing aptitude and excellent formation of tacky adhesive layer showing excellent pressure-sensitive adhesive characteristics with light irradiation of short time to consequently provide an adhesive agent with very high practical application features with possible assembly operation with very high productivity when said pressure-sensitive adhesive composition is used to assembly flexible printed circuit board.

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